Far-Infrared Spectroscopy of Thin Oxide Films

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INTRODUCTION

Our work at beamline 1.4.2 has involved the setting-up and use of Fourier-transform infrared (FTIR) spectroscopy in the range of 20-100 cm^{-1} to complement lower-frequency time-domain Terrahertz (TD-THz) studies of thin films of the oxides $SrRuO_3$ and $Bi_2Sr_2CaCu_2O_{8+\delta}$. We have demonstrated stable measurement in the desired frequency range, have characterized the optical properties of the $NdGaO_3$ and $LaAlO_3$ substrates on which the two films, respectively, were grown, and have obtained transmission data for $SrRuO_3$ which upon further analysis may help to illuminate the oxide's non-Fermi-liquid electrical properties.

SYSTEM CAPABILITIES

The largest portion of our work at beamline 1.4.2 this year has been the adaptation of the beamline's Bruker IFS 66v/S FTIR system to the configuration necessary for far-infrared measurements of thin films. We installed, aligned, and calibrated a Janis cold-finger cryostat, on which the thin-film samples are mounted for measurement. This cryostat allows us to control the samples' temperatures in the range of approximately 9 K to 300 K. We aligned the special beamsplitters (Mylar of thickness 23 μm and 50 μm) necessary for far-IR measurements, installed a power supply, and tested the operation of an Infrared Laboratories bolometer, a LHe-cooled infrared detector with an optical low-pass filter at 100 cm^{-1}. We found that the system provides data with an excellent signal-to-noise ratio for frequencies of 20-100 cm^{-1}; see figure 1. Moreover, a single beamsplitter (23 μ) provides data over the entire range, so that a sample's far-IR transmission can be determined in one experiment.
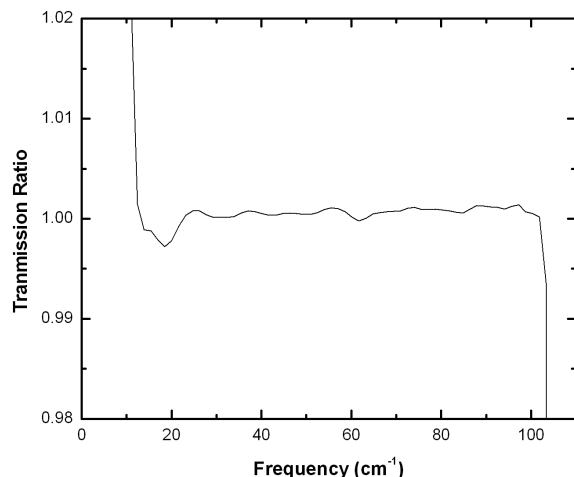

Figure 1: Measurement stability. The curve shown is the ratio of two consecutive sets of data, each collected for approximately 1 minute, with the source and external conditions held constant. The smallness of the deviations from unity shows excellent measurement stability in the range 20-100 cm^{-1}. The deviation below that range arises from opacity of the beamsplitter, and that above arises from a low-pass filter.

CHARACTERIZATION OF SUBSTRATES

Because thin films are grown on substrates, the optical transmission of a film must be normalized by that of the substrate. We measured the absolute transmission of the $NdGaO_3$ and $LaAlO_3$ substrates on which the $SrRuO_3$ and $Bi_2Sr_2CaCu_2O_{8+\delta}$ films, respectively, were grown; the results
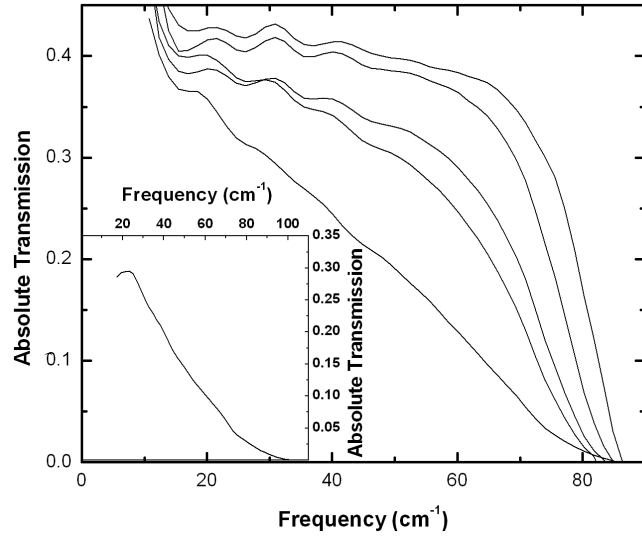
appear in figure 2. We found that both substrates are opaque at frequencies above ~ 100 cm^{-1}; measurement using an unfiltered detector showed that this opacity is intrinsic to the substrates, and not an artifact of the bolometer's low-pass filter. This opacity limits the possible bandwidth of optical measurements of films grown on these substrates.

Figure 2: Infrared transmission of NdGaO₃ substrate. From top to bottom: T = 9 K, 65 K, 110K, 150 K, 300 K. The curves for T < 300 K were fit to the form of the transmission arising from a single-oscillator absorption at approximately 90 cm^{-1}, as described in the text. **Inset**: The transmission of the LaAlO₃ substrate at T = 300 K is shaped similarly to that of NdGaO₃. The transmission of LaAlO₃ rises above 1000 cm^{-1} (not shown).

Our measurements of a film yield the ratio, T, of the transmission of the film-substrate complex to that of the substrate alone; we seek to fit the measured T to that arising from a theoretical form for the complex electrical conductivity, σ, of the film. In the thin-film approximation, for a substrate of index of refraction n, T is given by:

$$T = |(n+1)/(n+1+\sigma dZ)|^2$$

where d is the thickness of the substrate, and Z is 377 Ω. In order to perform the desired fit both the real and imaginary parts of n must be known, but both cannot be determined from measurements of optical transmission over a finite frequency range. However, we found that the measured transmission of NdGaO₃ (fig. 2) fits well to the form arising from a single-oscillator absorption at approximately 90 cm^{-1}, particularly for the lower temperatures; only the width of the oscillator was found to vary with temperature. We thus calculated the full complex n from the single-oscillator model.
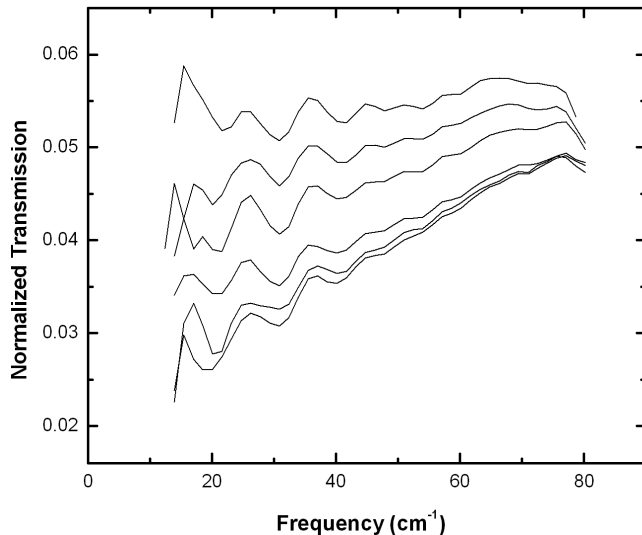
CHARACTERIZATION OF SrRuO₃ FILMS

Figure 3: Infrared transmission of SrRuO₃ film, normalized by the transmission of the substrate. From top to bottom: T = 65 K, 55 K, 45 K, 35 K, 25 K, 9 K. These curves are being fit to a form arising from non-fermi-liquid conductivity (see text). It is expected that the temperature dependence of the infrared transmission will reflect the temperature dependence of the inverse scattering rate, τ, which was determined by TD-THz methods.

The electrical properties of SrRuO₃ have attracted attention [1,2] because of the material's magnetic properties [3,4], and because of its similarities to the cuprate high-T_c superconductors. Recent measurements in the range 100-1000 cm^{-1}

[1] suggest non-Fermi-liquid electrical behavior, a behavior also thought to exist in the cuprates. We have measured the transmission of $SrRuO_3$ films at various temperatures; some of these data appear in figure 3. We are in the process of fitting these curves to a form for the transmission which arises from non-Fermi-liquid conductivity, σ, suggested by Ioffe and Millis [5].

A critical aspect of this fitting process is the connection with TD-THz data taken at lower frequencies. The inverse scattering rate τ, a parameter of the electrical conductivity, is given by $\tau = (d/d\omega)\,(\sigma_i/\sigma_r)$, where ω is the frequency, and σ_r and σ_i are the real and imaginary parts of σ, respectively. Since TD-THz measurements yield both σ_r and σ_i, our collaborators [6] have been able to accurately determine τ for temperatures below 70 K. It is expected that the temperature dependence of τ will be consistent with the temperature dependence of our far-infrared data in a non-Fermi-liquid model of the conductivity.

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